

Fabrication and characterization of GaN/InGaN/AlGaIn double heterostructure LEDs and their application in luminescence conversion LEDs (LUCOLEDs)

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Abstract

We report on the fabrication as well as on the optical and electrical characterization of violet and blue GaN/InGaN/AlGaIn double heterostructure light emitting diodes (DH LEDs) covering the 385nm-430nm spectral range. MOCVD grown epitaxial layer sequences were processed into mesa diodes by chemically assisted ion-beam etching and contact metallization. To achieve packaged LED devices the diode chips were encapsulated in transparent epoxy resin using standard technology.

Based on blue emitting diodes as primary light sources, white luminescence conversion LEDs (LUCOLEDs) have been fabricated. Using commercially available perylene dyes or YAG:Ce phosphors as the luminescent material, the LED radiation is converted into light of longer wavelengths by luminescence down-conversion (Stokes shift). In contrast to conventional LEDs which only emit quasi-monochromatic light, light of nearly all colors can be generated by this technique. By mixing the primary blue light with the radiation emitted from the converting material, also white and mixed colors have been generated.

Keywords: GaN, LED, luminescence conversion, white LED

1. Introduction

Wide bandgap (AlGaIn)N-based heterostructures are of considerable current interest for the realization of optoelectronic devices operating in the UV-to-green spectral range. The commercialization of high brightness blue and green light emitting diodes as well as the realization of violet emitting laser diodes have established (AlGaIn)N as an important material system for optoelectronic devices [1,2]. These short-wavelength emitting primary light sources can be used as efficient pumps to excite organic or inorganic luminescent materials for subsequent photon emission at lower energies. Mixing primary and luminescence light, a great variety of colors including mixed colors and white light can be generated [3,4,5].

In this paper we describe the development of violet, blue and green emitting GaN/InGaN/AlGaIn DH LEDs grown by low pressure MOCVD and report on the realization of luminescence conversion LEDs (LUCOLEDs) emitting white, green or red light.

2. GaN/InGaN/AlGaIn double heterostructure LEDs

2.1. MOCVD Growth

The epitaxial layer sequences required for a GaN/InGaN/AlGaIn DH LED (Fig. 1) were grown on c-plane oriented 2"-sapphire substrates in an AIXTRON AIX 200/HT low pressure MOCVD-reactor. After annealing the substrate at 1030°C a 25nm thin GaN nucleation layer was deposited at 530°C. $\text{Al}_y\text{Ga}_{1-y}\text{N}$ ($0 \leq y \leq 0.16$) layers were grown at 1030°C while $\text{In}_x\text{Ga}_{1-x}\text{N}$ films were deposited at temperatures between 680°C and 800°C. Silicon doping has been achieved without the appearance of a degrading surface morphology up to Si concentrations in the mid 10^{19}cm^{-3} range. The highest mobilities, $450\text{cm}^2/\text{Vs}$ at a free electron concentration of $2 \times 10^{17}\text{cm}^{-3}$, were obtained for samples with the lowest deliberate Si doping level. Mg-doped GaN films with chemical concentrations in the range of $4 \times 10^{17}\text{cm}^{-3}$ to $2 \times 10^{20}\text{cm}^{-3}$ have been prepared. After a 10 min thermal annealing in ambient N_2 at 700°C the heavily doped ($\geq 10^{19}\text{cm}^{-3}$) samples became p-conducting with RT hole densities up to $5 \times 10^{17}\text{cm}^{-3}$ and mobilities up to $15\text{cm}^2/\text{Vs}$.

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As test structure for the LED active region, undoped or Si doped $\text{In}_{0.1}\text{Ga}_{0.9}\text{N}$ films have been grown on the top of $3\mu\text{m}$ thick $\text{GaN}:\text{Si}$ layers. Thin films with $d \leq 60\text{nm}$ reveal RT PL spectra which are dominated by a single peak close to the band gap $E_g(x)$ of the InGaN .

2.2. LED Fabrication

In order to demonstrate the good quality of our epitaxial layers we have grown a $\text{GaN}/\text{InGaN}/\text{AlGaIn}$ DH LED showing electroluminescence in the violet, and blue spectral range depending on the In concentration in the active layer. A schematic drawing of a mesa diode, prepared from the above $\text{GaN}/\text{InGaN}/\text{AlGaIn}$ layer sequence by optical lithography and reactive ion etching, is shown in Fig. 1. Light emitted by the diode can be collected either from the backside through the transparent substrate or from the top through the semi-transparent p-contact.

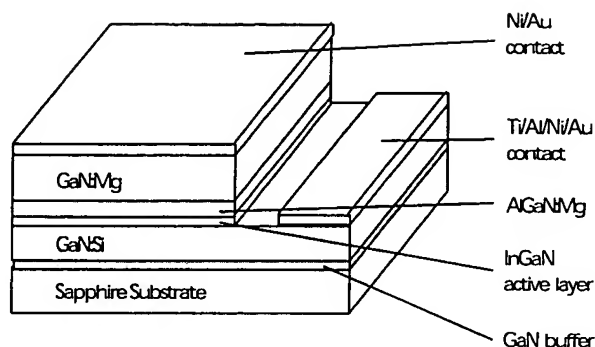


Fig. 1. Schematic drawing of a $\text{GaN}/(\text{InGa})\text{N}/(\text{AlGa})\text{N}$ double-heterostructure LED

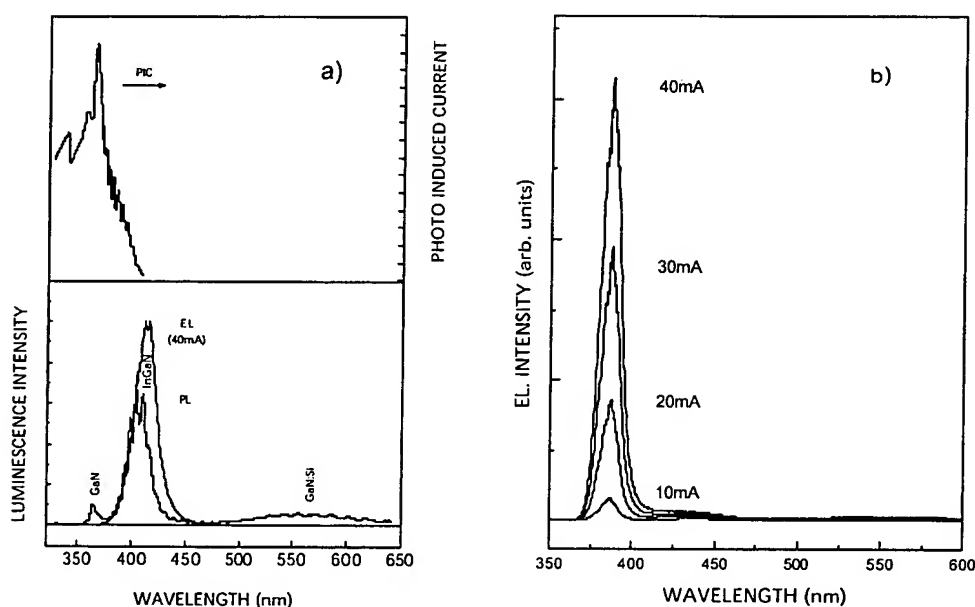


Fig. 2. (a) Emission and photoluminescence spectra of a deep blue (413 nm) emitting double heterostructure LED compared with the photoinduced current PIC, (b) Emission spectra of a violet (386 nm) emitting MQW LED at various currents as described in the text

The layer structure of the blue emitting DH LED consists of $3\mu\text{m}$ thick $\text{GaN}:\text{Si}$ ($[\text{Si}] = 2.5 \times 10^{18} \text{cm}^{-3}$) followed by 10nm $\text{In}_{0.1}\text{Ga}_{0.9}\text{N}:\text{Si}$, 15nm $\text{Al}_{0.12}\text{Ga}_{0.88}\text{N}:\text{Mg}$ ($[\text{Mg}] = 1 \times 10^{18} \text{cm}^{-3}$) and a $0.5\mu\text{m}$ thick p-type $\text{GaN}:\text{Mg}$ layer ($[\text{Mg}] = 2 \times 10^{19} \text{cm}^{-3}$). After annealing the layer system for 10min at 700°C in ambient N_2 , $300\mu\text{m} \times 300\mu\text{m}$ wide p-contacts ($10\text{nmNi} / 400\text{nmAu}$) were evaporated through a shadow mask. Apart from the contact area the epilayer material has been removed by reactive ion etching down to the $\text{n-GaN}:\text{Si}$. Subsequently $300\mu\text{m} \times 200\mu\text{m}$ wide

n-contacts ($15\text{nmTi} / 200\text{nmAl} / 40\text{nmNi} / 100\text{nmAu}$) separated by $400\mu\text{m}$ from the p-contacts have been evaporated through a shadow mask. Depending on the doping level no subsequent contact annealing has been necessary to achieve ohmic contacts. TLM measurements have revealed a specific contact resistance of about $5 \times 10^{-3} \Omega \text{cm}^2$ for p-contacts and $2 \times 10^{-5} \Omega \text{cm}^2$ for n-contacts. The processed structures have been bonded and housed using standard technology and their electroluminescence spectrum has been examined through the sapphire substrate.

The well defined emission band of the deep blue LED with a FWHM of 160 meV (22 nm) is peaked in the deep blue at about 3.0 eV, corresponding to an emission wavelength of 413 nm (Fig. 2a). At 30 mA, an output power of about 0.3 mW is obtained. The coincidence of the electroluminescence and photoluminescence bands with the onset of the photocurrent indicates that the luminescence is due to a near band transition in the InGaN layer. By decreasing the thickness of the active InGaN layer the emission peak is shifted further into the violet spectral range as seen for a MQW LED emitting at 386nm with a FWHM of 100 meV (11 nm) (Fig.2b). The emission peaks of both diodes do not shift with increasing current, which indicates the high material quality achieved. The turn-on voltage of the diode of about 3.5 eV is consistent with the band gap energy of the active region of the LED (Fig. 3).

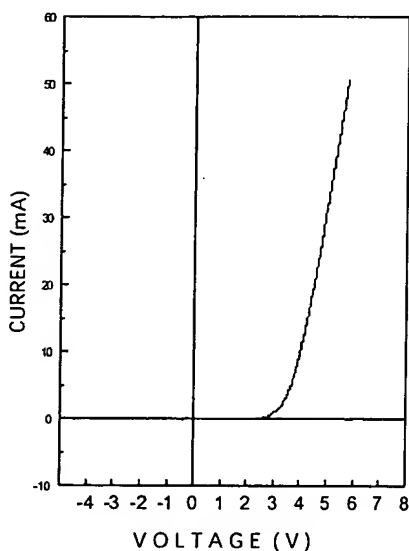


Fig. 3: Current voltage characteristic of a double-heterostructure LED emitting at 386 nm

3. Luminescence conversion LEDs

High-brightness GaN-based LEDs emitting in the blue, and in the future also in the UV spectral range, form the basis of luminescence converting LEDs (LUCOLEDs). The principle of the luminescence conversion is illustrated in Fig. 4. Here, a GaN LED chip mounted in a reflector cup is covered by a luminescence converter consisting of an organic luminescent dye dissolved in epoxy resin, while the whole system is embedded in transparent epoxy resin by standard LED technology.

When excited by the LED the organic dye (e.g. Lumogen BASF) totally absorbs the blue light and emits at longer

wavelength. In this way green, yellow and red emitting LEDs have been fabricated [3,4].

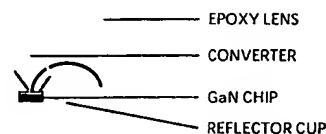


Fig. 4. Schematic structure of a GaN based luminescence conversion LED.

For incomplete absorption of the primary blue light by the LUCO material, also mixed colors can be generated by additive color mixing of the primary LED-blue with a secondary luminescence light. Most notably, additive color mixing of the primary LED-blue with secondary photoluminescence red results in magenta, a color not accessible by a single conventional pn-junction LED.

White emitting diodes can also be realized, if a green emitting and a red emitting dye are simultaneously added. The emission spectrum of such a white LUCOLED is composed of three spectral peaks, arising from the two different dye species and the blue LED [3].

3.1. YAG:Ce based LUCOLEDs

The principle of luminescence down-conversion of blue emitting LEDs is not restricted to organic luminescent materials. Also inorganic phosphor materials should be considered for LUCOLED applications since they show a superior UV and thermal stability. The situation encountered here is basically the same as found in fluorescent tubes.

For white LUCOLEDs the phosphor $\text{YAl}_3\text{O}_{12}:\text{Ce}^{3+}(4f^1)$, in short: YAG:Ce, is ideally suited, since the 4f - 5d transition of the Ce^{3+} ion absorbs in the blue spectral range while the corresponding emission is shifted into the yellow spectral range due to a pronounced Stokes-shift, caused by vibronic coupling of the excited 5d-level. For yellow light emission under blue photo-excitation, only one converter species is needed for white light generation [4,5], since the complementary colors blue and yellow result in white light after proper additive mixing (Fig. 5). The emission spectra of a white emitting YAG:Ce based LUCOLED are shown in Fig. 6 for three different drive currents. The two distinct emission bands from the GaN-based LED and the YAG:Ce converter

are clearly resolved at 417nm and at around 535 nm, respectively. The quantum conversion η_q as well as the power conversion η_p efficiencies of the YAG:Ce and Lumogen dye converters in LUCOLEDs are shown in Tab.1.

Table 1

Quantum η_q and power η_p conversion efficiencies of YAG:Ce and Lumogen dye converters in LUCOLEDs

Color	Converter	η_q (%)	η_p (%)
White	YAG:Ce L175	75	61
Green	Lumogen F 083	57	46
Red	Lumogen F 300	80	57

Changing the current over a wide range has no influence on the chromaticity of the LUCOLED ($x=0.31$, $y=0.34$) being close to the equal energy point.

A wide range of whitish colors can be realized by fabricating LUCOLEDs with different YAG:Ce concentration. Increasing this concentration shifts the color from blue to yellow. Their chromaticity coordinates are close to the straight line interconnecting the points of the blue pump and the yellow phosphor (Fig. 7). In this way, LUCOLEDs with different whitish hues can be realized.

4. Conclusions

High-brightness GaN-based LEDs emitting in the blue, and in the future also in the UV spectral range, form the basis of colored and white light emitting luminescence converting LEDs. For white LUCOLEDs there are wide fields of application, e.g. in the car industry for dashboard and indoor illumination or for display backlighting. The long lifetime (≈ 100000 h) of a LUCOLED, combined with its small size and low electric power consumption are important advantages. White LUCOLEDs based on YAG:Ce phosphors produced by Nichia are already on the market and Siemens will start the commercialization in 1998.

Acknowledgments

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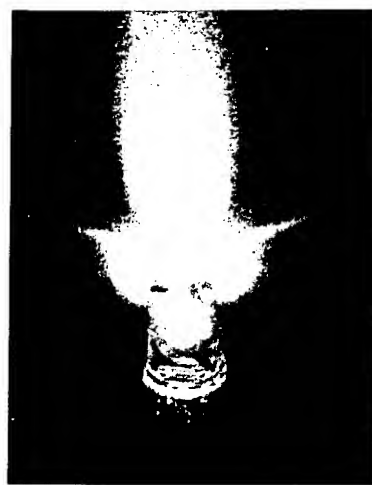


Fig. 5. Photograph of a white emitting YAG:Ce based LUCOLED

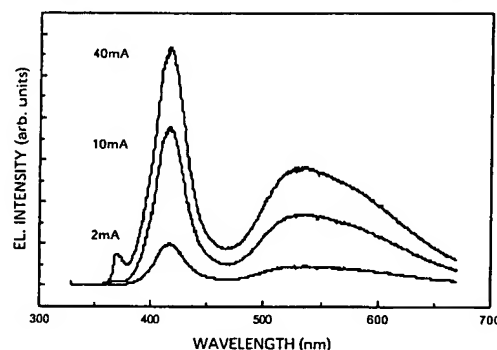


Fig. 6. Emission spectra of a white emitting YAG:Ce based LUCOLED at different currents

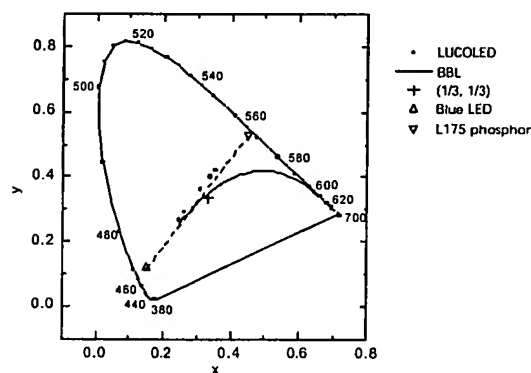


Fig.7. CIE chromaticity points of YAG:Ce based LUCOLEDs and of the corresponding GaN LED and YAG:Ce phosphor. The chromaticity points (marked by circles) of LUCOLEDs, produced by varying the YAG:Ce concentration, are on the straight line connecting the chromaticity points of the LED and the YAG:Ce phosphor.

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